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Abstract

We report the results of laser-induced damage and nonlinear absorption measured for CdTe and other selected II-VI materials. These studies were conducted using pulsed 1.06 µm radiation from a Nd:YAG laser. The laser pulsewidth was varied from approximately 40 psec to 9,000 psec (9 nsec). The laser-induced breakdown irradiance measured for CdTe over this pulsewidth range scaled as $t_p^{-1/2}$ [t_p is the laser pulsewidth (FWHM)]. Nonlinear phtoacoustic spectroscopy was used to monitor the nonlinear and linear absorption in the samples in which laser-induced damage thresholds were measured.

Introduction

The II-VI compounds and various single element semiconductors are used as window materials for continuous output lasers operating in the 10.6 µm region. These materials are also used as the high index of refraction component of multilayer dielectric coatings for laser and other infrared optics applications. While these materials have proved useful for nonlaser infrared applications and for many continuous output laser applications, they are generally avoided in pulsed laser systems.

There are a variety of fundamental reasons to avoid the high index materials for pulsed laser applications. The high index means very high fresnel losses at interfaces and the field enhancement associated with surface scratches and other defects[1,2] is more pronounced. Bettis, Guenther, and Glass[3] have argued that high index materials will have lower damage thresholds based upon local field considerations. Finally, Wang's rule[4] for nonlinear indices of refraction, n_2, implies that a material with a large linear index will also have a large n_2, and thus the problems associated with self-focusing will be most pronounced for high index materials.

Despite these problems there are a variety of pulsed laser applications which require the use of the II-VI compounds, e.g., a system that has optics which are shared by lasers of different frequencies and different pulsewidths. An example of such a system would be a pulsed Nd:YAG range finder used with a continuous output CO_2 laser. These materials are also of interest in integrated optics and phase conjugation; two applications for which a large nonlinear index of refraction is advantageous. In any such application it is important to know the operating limits set by laser-induced damage to the materials.

In this study we measured the pulsed laser-induced surface damage threshold of CdTe, ZnSe, CdS and ZnTe at 1.06 µm. The study emphasized CdTe because of its use as a pulse shaping device[5] in laser fusion systems and other applications and because it is a good "model system" for use in studying nonlinear absorption. The band gap in CdTe is 1.32 eV, which lies between 1.06 µm and 0.53 µm. The position of the band gap in CdTe means that it is transparent to 1.06 µm radiation but has a relatively large two-photon absorption coefficient. These experiments show that absorption in CdTe is dominated by two-photon absorption at 1.06 µm near the damage threshold. However, the surface damage threshold irradiance scales as $t_p^{-1/2}$ (t_p is the laser pulsewidth) which is characteristic of damage due to surface contamination.

Experimental

The laser source for the psec studies was a passively mode-locked, microprocessor-controlled[6] Nd:YAG laser system operating at 1.06 µm. A single pulse of measured Gaussian spatial and temporal intensity distribution was switched from the mode-locked train and amplified. The temporal pulsewidth was variable between 30 and 200 psec [full widths at half maximum (FWHM)] by selecting various etalons as the output coupler. The width of each pulse was monitored by measuring the ratio, R, of the square of the energy in the fundamental (1.06 µm) to the energy in the second harmonic, produced in a $LiIO_3$ crystal. This radio is directly proportional to the laser pulsewidth as long as the spatial profile remains unchanged.[7] The ratio was calibrated by measuring the pulsewidth using type-I second-harmonic autocorrelation scans. The observed three-to-one signal to-background ratios indicated clean mode locking.[8] To ensure that the ratio, R, is proportional to the pulse width and provides a valid pulsewidth monitor, scans were performed for all output coupler etalons.

The laser beam was focused onto the sample surface with a single element lens of "best form" design, i.e., designed for minimum spherical aberrations. The focal position (547 ± 2 mm from the lens mechanical center) was determined by a series of pinhole scans as a function of distance from the focusing lens. The radius of the unfocused beam was 2.35 mm, resulting in an f/232 imaging system. The focal radius at the $1/e^2$ point of the irradiance was 75 ± 3 μm as determined by pinhole scans. Figure 1 is a plot of the beam scan data and a Gaussian best fit to the data. Some of the data was taken with the sample at the focal position and some data was taken with the specimen 2.70 cm behind the focal point. The beam radius was calculated using Gaussian optics to be 139 ± 4 μm at the position 2.70 cm behind focus. The beam was not scanned at this position, however; it was scanned at 2.25 cm behind focus. The measured radius at this position was 122 ± 2 μm, which is within 5% of the 116 μm value predicted by Gaussian optics.

The beam irradiance was controlled by varying the voltage in the amplifier stage of the laser. Beam scans were conducted at various amplifier settings thereby verifying that the beam spatial profile was unchanged over the range of amplifier settings used. The pulse energy into the specimen and transmitted through the specimen was monitored for each laser shot.

A piezoelectric transducer was mounted on the samples using pressure contact. The transducer was used to monitor the acoustic signal generated in the sample by linear and nonlinear absorption of light by the sample. The optoacoustic technique used in this experiment is described elsewhere.[9,10] The optoacoustic signal and the transmission of the sample were monitored as the laser output was increased to a value which produced damage. In this experiment damage was taken to be any perceptable change in the sample as viewed with a 20x microscope.

A Q-switched Nd:YAG laser operating at 1.06 μm was used to measure the damage threshold of the CdTe specimen. The pulsewidth for this device was 9 nsec (FWHM) as measured using a high speed photodiode read by a one GHz bandwidth oscilloscope. The focal radius of the Q-switched beam at the sample surface was 26 μm as calculated using Gaussian optics and the measured unfocused beam parameters. The laser system is more completely described elsewhere.[12]

The CdTe, CdS and ZnTe samples were single crystals. The ZnSe specimen was polycrystalline material grown by chemical vapor deposition by the Raytheon Research Laboratories, Bedford, Mass.

Results and Discussions

The results of the psec surface damage measurements are summarized in Table I. The threshold values given are the irradiance levels, I_B, which produce damage 50% of the time as determined using the procedure described by Porteus et al.[12] In all the psec measurements the pulsewidth of each shot was measured and the irradiance calculated. Only shots within the pulsewidth range shown in Table I were used in determining the damage thresholds.

The laser-induced damage observed in this work always occurred on the front or entrance surface. The imaging lens for the psec measurements was used at f/232. This means that the depth of focus was very large and thus the beam radius was approximately the same at the front and exit surfaces. For this situation (neglecting absorption) the local electric field of the light beam at the rear surface is larger than that at the front surface by a factor $2n/n+1$ where n is the index of refraction of the specimen.[13] Thus, the exit surface should fail at irradiance levels $4n^2/(n+1)^2$ lower than the front surface. For CdTe this factor is approximately 2.3 and thus the exit surface should fail at irradiance levels equal to approximately 0.43 times the front surface damage threshold.

The fact that front surface damage preceeds exit surface damage in these experiments can be understood in terms of the nonlinear absorption which preceeds damage in the materials studied. Figure 2 is a plot of the inverse of the transmission as a function of incident irradiance for CdTe. The laser pulsewidth for this data was approximately 40 psec. The points are actual data and the solid line the theoretical prediction based on two-photon absorption.[17] The intercept (zero intensity) corresponds to the fresnel losses and small linear absorption losses. Note that the transmission at ~500 MW/cm^2 is down by a factor of 5 or 2.5 times the loss due to fresnel reflections and linear absorption. This means that for intensities of 500 MW/cm^2 and above, the field enhancement at the exit surface will be negated by the high nonlinear absorption. Note that the measured damage threshold for the front surface was 2.0 GW/cm^2 for this pulsewidth and thus the nonlinear absorption reduced the flux at the rear surface to a value below the measured front surface threshold.

The plot in Fig. 2 shows that for relatively low intensities, i.e., below about 400 MW/cm^2, the nonlinear absorption fits the theoretical prediction for two-photon absorption. The data starts to deviate significantly at higher intensities. One possible explanation is that free carrier absorption by the two photon generated carriers becomes important.[14,15,16] The presence of free carriers can further reduce the intensity at the rear surface by plasma defocusing.[17,18,19]

Nonlinear transmission and nonlinear optoacoustic measurements were made for all the materials listed in Table I. Absorption in CdTe and ZnTe was dominated by two-photon absorption and absorption in CdS and ZnSe was dominated by three-photon absorption. The analysis of this nonlinear absorption data is not yet complete and will be published at a later date. While the absorption in these specimens was dominated by multiphoton processes, damage appeared to be due to linear absorption caused by surface defects and/or surface contaminaion as the following analysis shows.

Material	Surface Damage Threshold Irradiance (GW/cm^2)		
	Focal Radius ($1/e^2$ Point of the Irradiance)		
	139 μm	75 μm	26 μm
CdTe	a) 1.24 ± .04 t_p = 124 ± 10 ps b) 1.18 ± .06 t_p = 108 ± 10 ps c) 2.06 ± .10 t_p = 45 ± 5 ps	a) .82 ± 0.03 t_p = 188 ± 15 ps b) 1.96 ± 0.10 t_p = 42 ± 3ps	0.10 ± 0.01 t_p = 9000 ± 500 ps
ZnTe	0.43 ± 0.03 t_p = 46 ± 5 ps		
ZnSe		a) 12.4 ± 0.9 t_p = 41 ± 3 ps b) 14.9 ± 1.5 t_p = 34 ± 3 ps	
CdS		20.0 ± 0.6 t_p = 35 ± 3 ps	

Table I. Summary of 1.06 μm Surface Damage Threshold Data. The top number in each entry above is the damage threshold irradiance in units of GW/cm^2.

The data in Table I indicates that the surface damage threshold for CdTe with 40 psec pulses is independent of the focal radius to within the uncertainty of the measurement. Bettis, House, and Guenther[20] have used a variety of surface damage threshold data to derive a sealing law which says that the surface damage threshold irradiance of dielectrics scales as ~ 1/w where w is the focal radius. However, their model is based on the spatial spreading of a laser-induced plasma and no plasma was observed in these experiments. Examination of the damage sites and the observed pulsewidth dependence both indicate that damage to these materials was thermal in origin and associated with surface defects or contamination. We attribute the lack of spot size dependence to the fact that surfaces have a relatively high density of defects which initiate the damage and thus the probability of finding a defect within the beam radius is essentially unity over the smallest spot size used (26 μm radius).

The CdTe data in Table I is plotted in Fig. 2. The solid line in Fig. 2 is a least squares fit of the data to a $t_p^{-1/2}$ dependence. Note that the fit to the data is very good over the entire 40 to 9000 psec range. The I_B dependence on the laser pulsewidth t_p can be understood in terms of a simple thermal failure model. Suppose that there is a thin absorbing layer on the surface and damage is initiated by raising the temperature of this layer to a value which results in melting or an irreversible phase change which changes the surface appearance. If such a layer has thickness δ, less than the thermal diffusion depth, i.e., $\delta < v_s t_p$ where v_s = velocity of sound in the material and t_p is the laser pulsewidth, then heat is lost from the irradiated area during the laser pulse. The net result is that more energy is required to raise the surface temperature a given amount for relatively long pulses than for relatively short pulses. One dimensional heat transfer calculations (diffusion along the radius of these relatively large spots can be ignored) indicate that the threshold energy density should scale as $t_p^{1/2}$ and the threshold irradiance should scale as $t_p^{-1/2}$.

Sparks and Duthler[21] have modeled thermal damage due to surface inclusions. Their solution to the heat transport equation for the case of a metallic inclusion for which the absorption skin depth is much less than the thermal diffusion depth predicts that $I_B \sim t_p^{-1/2}$. Their model assumes that the skin depth is much less than the inclusion radius and treats absorption by spherical inclusions the same as absorption by a plane-slab with a δ-function source. Thus, this model should work equally well for absorption by a thin contamination layer (such as an oxide layer 10's to 100's of angstroms thick). The fact that the $t_p^{-1/2}$ dependence is seen for pulses as short as 42 psec indicates that the absorption which leads to damage occurs in a layer of thickness $\delta \lesssim 2000$ Å thick. Such absorption could be due to metallic inclusion or a thin surface contamination layer. Experiments are planned to test this theory by measuring the pulsewidth dependence of chemically etched surfaces.

A final observation regarding these measurements is that we observed a visible flash in the CdS and ZnSe sample prior to damage. This visible light was emitted in the forward direction and was fairly well collimated. Spectral analysis of the emission from ZnSe revealed that the light was monochromatic and the wavelength was within ±1 Å (the limit of our spectrometer resolution) of frequency doubled 1.06 μm, i.e., 0.5320 μm. The ZnSe sample was polycrystalline with random crystallite orientation and the observed second harmonic generation was angle insensitive through relatively inefficient. The threshold for visual detection (viewed through 1.06 μm laser safety goggles) of the second harmonic from the ZnSe was approximately 4 MW/cm^2. This is substantially below the intensity required to burn polaroid film. The relatively low threshold intensity for the generation of the second harmonic and the angle insensitivity make CVD ZnSe a relatively cheap, easy to use frequency doubling material where high efficiency is not needed and a practical device for tracking and monitoring the presence of 1.06 μm beams.

Summary

The laser-induced surface damage threshold was measured for the front surface of CdTe at a variety of pulsewidths and focal spot radii at 1.06 μm. The results indicate that the damage threshold irradiance is independent of the focal radius and scales as $t_p^{-1/2}$. The observed surface damage in these II-VI materials appears to be caused by linear absorption by surface defects and/or surface contaminants. Two-photon absorption was determined the dominant absorption mechanism for CdTe and ZnTe and three-photon processes were dominent for ZnSe and CdS. The depletion of the beam due to these nonlinear absorption processes prevented rear surface damage in all the specimens studied.

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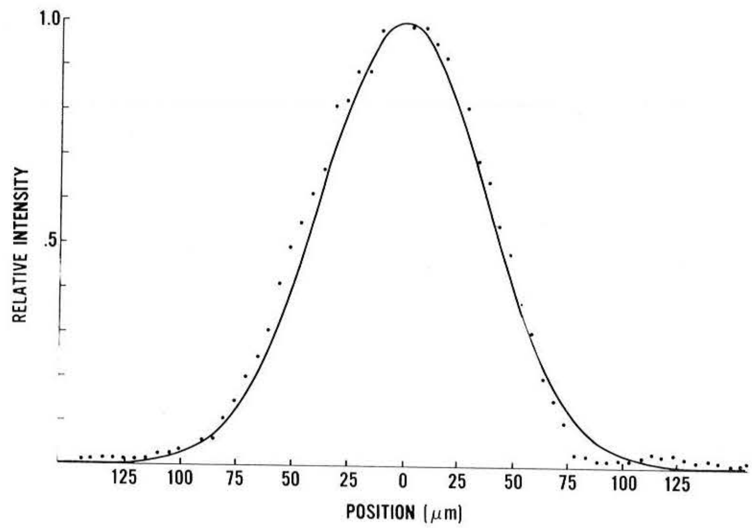

Fig. 1 Pinhole scan of the focused beam from the psec laser. The dots are the actual data points and the solid line is an ideal Gaussian of width 77 μm. After correction for the finite size of the pinhole used to make this scan the focused beam radius was determined to be 75 ± 3 μm.

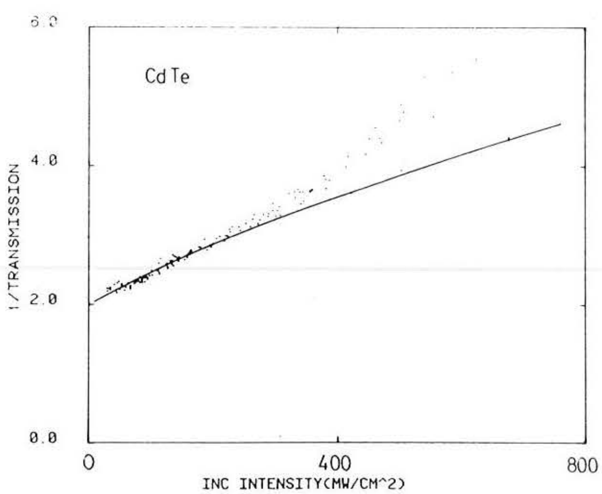

Fig. 2 Nonlinear absorption in CdTe. Here the inverse of the transmission (the ratio of the input beam to the transmitted beam) is plotted as a function of the input irradiance. The solid line is the theoretical curve for two-photon absorption neglecting two-photon induced free hole absorption.[14]

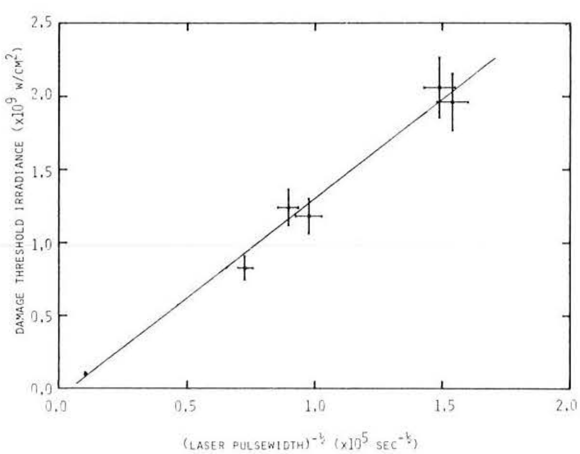

Fig. 3 Front surface damage threshold irradiance for CdTe vs. $t_p^{-1/2}$. The irradiance is given in GW/cm^2 and $t_p^{-1/2}$ is in units of 10^5 $sec^{-1/2}$. The solid line is the lease squares fit of the data to a $t_p^{-1/2}$ dependence (t_p is the laser pulsewidth, FWHM). All the data was taken with Nd:YAG laser operating at 1.06 μm.